

Event Driven Langevin simulations of Hard Spheres

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The blossoming of interest in colloids and nano-particles has given renewed impulse to the study of hard-body systems. In particular, hard spheres have become a real test system for theories and experiments. It is therefore necessary to study the complex dynamics of such systems in presence of a solvent; disregarding hydrodynamic interactions, the simplest model is the Langevin equation. Unfortunately, standard algorithms for the numerical integration of the Langevin equation require that interactions are slowly varying during an integration time-step. This is not the case for hard-body systems, where there is no clearcut between the correlation time of the noise and the time-scale of the interactions. Starting first from a splitting of the Fokker-Plank operator associated with the Langevin dynamics, and then from an approximation of the two-body Green's function, we introduce and test two new algorithms for the simulation of the Langevin dynamics of hard-spheres.

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I. INTRODUCTION

Hard spheres (HS) are a reference system for structural and dynamical theories of fluids [1, 2], but idealized: the infinitely steep potential is essentially a way of capturing the effects of steric interactions. On the atomic or the molecular scale two body interactions are mostly modelled by Lennard-Jones or Coulomb potentials; experiments on colloids shift the length scales of interest up to roughly 1 nm to 1000 nm where objects can behave as hard bodies and are still small enough to exhibit thermal or Brownian motion in a solvent. Dynamical light scattering [3, 4] has already provided a rich collection of data for such systems, encouraging a considerable effort in understanding the dynamics; the possibility of following single particle trajectories via confocal microscopy of latex particle [5] has allowed a direct view on an experimental realization of HS systems and their dynamics [6, 7].

The simplest model of a suspension of neutral particles is to consider a system of HS in an ideal solvent with no hydrodynamic interactions; real suspensions are often described in terms of their deviations from such ideal system. This is the most interesting model for theoreticians and many results have been derived: the two body case (and hence the low density case) has been solved exactly [8, 9], while at moderate and high packing fractions various Enskog-like [10, 11] or Mode Coupling theories [12, 13] have been applied to understand the dynamics. While hydrodynamic interactions (HI) are well understood at low particle densities, much less is known at high densities, and theories often proceed by disregarding them [14]. As an example, theories regarding glass transition often disregard HI effect, like in the case of the Mode Coupling theory for Brownian hard spheres [13, 15, 16] or Brownian hard discs in shear flow [17].

Non-HI simulations therefore have their place in testing such theories, and in circumventing the huge effort needed to simulate HI [18–21].

In order to validate non-HI theories for HSs it is necessary to use computer simulations, as only a qualitative agreement is to be expected among non-HI theories and data for real suspension. Standard simulation methods for Brownian dynamic like the well-known Ermak-McCammon [22] require continuous potentials; to circumvent such problem several algorithms have been introduced with various degrees of justification [23–26] for the over-damped dynamics; only recently it has been recognized that in the case of hard interactions such simulations are better performed by event-driven (ED) codes [27–29]. We introduce two new ED algorithms that go beyond the over-damped approximation and allow for the simulation of the full Brownian dynamics of HSs.

II. METHODS

We consider a system of N HSs governed by the Langevin equation

$$\begin{cases} \partial_t \mathbf{v}_i = -\gamma \mathbf{v}_i + \mathbf{a}_i + \boldsymbol{\xi}_i \\ \partial_t \mathbf{r}_i = \mathbf{v}_i \end{cases} \quad (1)$$

for the positions \mathbf{r}_i and the velocities \mathbf{v}_i ; here γ is the friction constant, $\mathbf{a}_i = -m^{-1} \partial_{\mathbf{x}} U$ the acceleration, m is the mass of the HSs, U is the potential energy and $m\boldsymbol{\xi}_i$ are the zero-mean random forces due to the solvent. We assume that such random forces are delta correlated and satisfy the fluctuation-dissipation theorem

$$\langle \boldsymbol{\xi}_i(\mathbf{x}, t) \otimes \boldsymbol{\xi}_j(\mathbf{x}', t') \rangle = \gamma \frac{2k_B T}{m} \delta(\mathbf{x} - \mathbf{x}') \delta(t - t') \delta_{ij} \mathbf{1} \quad (2)$$

In the case of continuous interactions, it is possible to define stochastic Taylor expansions [30]; correspondingly, integration schemes of the k -th order with errors of order $(\Delta t)^k$ in the time-step Δt can be introduced [31]. In the case of hard-body interactions, all the standard machinery of stochastic calculus breaks down due to the singular

nature of the interaction potential and new methods must be developed.

We consider the Fokker-Plank equation associated to the SDE (1) (Kramers' equation [32])

$$\partial_t W = \mathbf{L}_K W \quad (3)$$

where $W(\mathbf{r}, \mathbf{v}, t)$ is the probability distribution function (PDF) for the positions $\mathbf{r} = \{\mathbf{r}_i\}$ and the velocities $\mathbf{v} = \{\mathbf{v}_i\}$ of the particles, $v_{th}^2 = k_B T/m$ relates to the temperature and

$$\mathbf{L}_K = \gamma (\partial_{\mathbf{v}} \cdot \mathbf{v} + v_{th}^2 \partial_{\mathbf{v}}^2) - (\mathbf{v} \cdot \partial_{\mathbf{r}} + \mathbf{a} \cdot \partial_{\mathbf{v}}) \quad (4)$$

is the Kramer operator. Integrating the SDE (1) for a finite time-step Δt corresponds to extracting a configuration $\{\mathbf{r}^{t+\Delta t}, \mathbf{v}^{t+\Delta t}\}$ according to the probability $e^{\mathbf{L}_K \Delta t} \delta(\mathbf{x} - \mathbf{x}^t, \mathbf{v} - \mathbf{v}^t)$.

III. SPLITTED BROWNIAN DYNAMICS

To obtain a numerical approximation, a powerful approach is to split the evolution operator $e^{\mathbf{L}_K \Delta t}$ in a product $e^{\mathbf{L}_K \Delta t} \approx \prod_i e^{a_i \mathbf{L}_i \Delta t}$ of exactly-integrable operators \mathbf{L}_i

[33] ensuring that that the decomposition is positive (i.e. all $a_i > 0$) [34]. Therefore, to each splitting corresponds an algorithm in which in a single time-step Δt the operators $e^{a_i \mathbf{L}_i \Delta t}$ are applied in sequence.

We first choose to split \mathbf{L}_K into the reversible (or streaming) operator $\mathbf{L}_{rev} = -(\mathbf{v} \cdot \partial_{\mathbf{r}} + \mathbf{a} \cdot \partial_{\mathbf{v}})$ and the irreversible (or collision) operator $\mathbf{L}_{irr} = \gamma (\partial_{\mathbf{v}} \cdot \mathbf{v} + v_{th}^2 \partial_{\mathbf{v}}^2)$ [35]; we indicate the corresponding algorithm as Splitted Brownian Dynamics (SBD).

The operator \mathbf{L}_{rev} is the Liouvillian associated to the Hamiltonian $\mathcal{H} = m \mathbf{v} \cdot \mathbf{v}/2 + U$. In the case of step potentials, the associated reversible equation of motion can be integrated via event-driven molecular dynamics (EDMD) [36] with a precision limited only by the numerical round-off errors; therefore the propagator $e^{\mathbf{L}_{rev} \Delta t}$ can be implemented with extreme accuracy.

The operator \mathbf{L}_{irr} corresponds to the interaction with the bath; the associated SDE is $\partial_t \mathbf{v} = -\gamma \mathbf{v} + \xi$ can be exactly integrated giving an explicit formula for the evolution $\mathbf{v}^{t+\Delta t} = e^{\mathbf{L}_{irr} \Delta t} \mathbf{v}^t$:

$$\mathbf{v}_{i,\alpha}^{t+\Delta t} = e^{-\gamma \Delta t} \mathbf{v}_{i,\alpha}^t + \sqrt{v_{th}^2 (1 - e^{-2\gamma \Delta t})} \Gamma \quad (5)$$

where Γ is a unitary Gaussian random variable and $\alpha \in \{x, y, z\}$.

The algorithm for the single SBD time-step $e^{\mathbf{L}_{rev} \Delta t} e^{\mathbf{L}_{irr} \Delta t}$ consists therefore in an EDMD simulation [36] of length Δt followed by a thermalization of the velocities according to eq.(5). We notice that the error is at most quadratic (as can be checked via Taylor expansion $e^{\mathbf{L}_{rev} \Delta t} e^{\mathbf{L}_{irr} \Delta t} = e^{\mathbf{L}_K \Delta t} + \mathcal{O}(\Delta t^2)$) and *regards only in the dynamics*; in fact, SBD is equivalent (upon identifying the angle α mixing reversible and

irreversible evolution with $\cos(\alpha) = e^{-\gamma(t-t')}$) to the Generalized Hybrid Monte Carlo [37] and therefore explores the canonical ensemble as long as the propagation steps $e^{\mathbf{L}_{rev} \Delta t}, e^{\mathbf{L}_{irr} \Delta t}$ can be exactly implemented (as in our case).

It is therefore of interest to give some physical bounds on the magnitude of the feasible time-step Δt . First, we notice that for $\Delta t \rightarrow \infty$ the dynamics reduces to and MD simulations where velocities are extracted each Δt from a Maxwellian; therefore if the time-step is much bigger than the average inter-particle collision time, results of classical MD are to be expected. Accordingly, we find that for big Δt the algorithm overestimates the diffusion coefficient (fig. 1); this is to be expected as the mean free path (in absence of collisions) of a particle is of order $v_{th} \Delta t$ instead of $\gamma^{-1} v_{th} \sqrt{\Delta t}$. Second, the magnitude of Δt is naturally bounded the damping time $\tau = \gamma^{-1}$; therefore the SBD is not well indicated for simulations in the over-damped limit $\gamma/m \rightarrow \infty$. Accordingly, we find that SBD overestimates diffusion coefficients for $\Delta t \gtrsim \gamma^{-1}$ (fig.1); it is therefore necessary to develop an alternative approach for the simulation of systems with high damping.

IV. APPROXIMATE GREEN'S FUNCTION DYNAMICS

It has been shown in [29] that the over-damped limit of eq.(1) can be simulated efficiently using ED codes[29]. The algorithm relies on considering time steps Δt small enough so that mostly binary collisions are relevant, i.e. the average displacement should be less than the average inter-particle separation. Moreover, average displacement should be smaller than the HSs' radii in order to map the interaction of two nearby HSs in the problem of a random walk near a reflective wall. Under such approximations, the true two-body stochastic dynamics for over-damped Brownian HSs can be implemented by algorithm of [29] in which each step consists in predicting the displacements $\Delta \mathbf{x}$ of the HSs via the free propagator, introducing fictive velocities $\mathbf{v}^f = \Delta \mathbf{x}/\Delta t$, and performing an EDMD with such fictive velocities during t and $t + \Delta t$. We extend such approach to the general Brownian case.

First, we need to predict the positions of the HSs after a time-step Δt according to their free propagation, i.e. the solution of eq.(1) with no interaction ($\mathbf{a} = 0$):

$$\begin{cases} \mathbf{v}^{t+\Delta t} = \mathbf{v}^t + \overline{\Delta \mathbf{v}} + \Delta \mathbf{v}_R \\ \mathbf{r}^{t+\Delta t} = \mathbf{r}^t + \overline{\Delta \mathbf{r}} + \Delta \mathbf{r}_R \end{cases} \quad (6)$$

The particle displacements contain both systematic parts $\overline{\Delta \mathbf{v}} = (e^{-\gamma t} - 1) \mathbf{v}^t$, $\overline{\Delta \mathbf{r}} = \gamma^{-1} (1 - e^{-\gamma t}) \mathbf{v}^t$ and stochastic displacements. The stochastic displacements $\Delta \mathbf{v}_R$, $\Delta \mathbf{r}_R$ are zero-mean correlated gaussian variables with variances $\langle \Delta \mathbf{v}_R^2 \rangle = m^{-1} k_B T (1 - e^{-2\gamma t})$, $\langle \Delta \mathbf{r}_R^2 \rangle = \gamma^{-1} m^{-1} k_B T [2t - \gamma^{-1} (3 + 4e^{-\gamma t} + e^{-2\gamma t})]$ and cross-correlation $\langle \Delta \mathbf{r}_R \Delta \mathbf{v}_R \rangle = \gamma^{-1} m^{-1} k_B T (1 - e^{-\gamma t})^2$ [38].

If we consider a time-step such that the average displacement is less than the average inter-particle separation, we can consider only the corrections due to two-body interactions. In the limit of small Δt , a couple of HSs will interact only when they start from nearby positions. In particular, if $\gamma^{-1}v_{th}\sqrt{\Delta t} \ll \sigma$, i.e. the average free displacement is much smaller than the diameter σ of the HSs, the dynamics of two particles A and B can be approximated as the Langevin dynamics of a point particle at a distance $(\mathbf{r}_A - \mathbf{r}_B)(1 - \sigma/|\mathbf{r}_A - \mathbf{r}_B|)$ from a flat wall. It is possible to solve such problem with a straightforward generalization of the image method applied in [29]. In fact, the solution given by the free particle Green's function plus an image particle with a reflected velocity beyond the reflective wall (fig. 2) correctly satisfies the zero-current boundary condition $\hat{\mathbf{n}} \cdot \mathbf{j}|_{wall} = 0$, where $\hat{\mathbf{n}}$ is the normal to the wall and $\mathbf{j}(\mathbf{r}, t) = \int \mathbf{v} W(\mathbf{r}, \mathbf{v}, t) d\mathbf{v}$ is the probability current for the position.

Such solution can be implemented exactly by predicting the new positions and velocities $\mathbf{r}^{t+\Delta t}, \mathbf{v}^{t+\Delta t}$ according to eq.(6), defining fictive velocities $\mathbf{v}^f = (\mathbf{r}^{t+\Delta t} - \mathbf{r}^t)/\Delta t$ and performing an EDMD simulation with such fictive velocities during Δt ; if a collision happens, the component of the relative velocity normal to the contact point must be reflected for both the fictive \mathbf{v}^f and the predicted velocities $\mathbf{v}^{t+\Delta t}$. We indicate such algorithm as the approximate Green's function dynamics (AGD). In the over-damped limit, the prediction of the velocities and positions decorrelates and the algorithm correctly reduces to the over-damped case of [29].

As for the SBD algorithm, it can be proven that the AGD scheme respects detailed balance and ergodicity and therefore explores the correct ensemble for HSs; hence, errors are again only in dynamic quantities. At difference with SBD, we have no analytic estimate for the error; nevertheless, we expect that the mean-free path in absence of collisions $\gamma^{-1}v_{th}\sqrt{\Delta t}$ must be smaller than the radius of the HSs in order to satisfy the flat-wall approximation, and must be smaller than the average inter-particle distance in order to avoid multiple collisions (hence higher than two-body effects) during Δt .

In order to check that the behaviour of AGS is driven just by geometrical considerations, we have simulated HS systems at different γ and ϕ varying the time-step Δt in the range $[10^{-2}, 10^0]$ (reduced units). At difference with SBD where diffusion can vary even by a order of magnitude in such a Δt range, the values of D measured from AGD vary a few percent over the range and long simulations are been necessary to have enough statistics to detect the behaviour of D that would otherwise look flat. In fig. 3 we show that the measured diffusion coefficient D versus the AGS simulation time-step displays a plateau (i.e. fluctuations become much smaller than 1%) already for $\Delta t \lesssim 0.1$ regardless of γ and ϕ .

V. CONCLUSIONS

Hard spheres, and in general hard body systems in suspension, have become a realistic model due to the developments of experimental techniques for the investigation of colloidal systems and nano-particles; yet the dynamics of such systems is hard to simulate via the standard Brownian dynamics algorithms. In fact, classical continuous-time algorithms fail due to instantaneous character of the interactions; we have shown instead how it is possible to simulate the full Langevin dynamics of Hard Spheres.

First, we have shown how the simplest splitting of the stochastic evolution operator (a technique often referred to as "Trotterization" from Trotter's seminal work[39]) allows to write an algorithm (the Splitted Brownian Dynamics - SBD). The SBD algorithm becomes inefficient of high viscosities but via the operator-splitting technique could easily take account for the interaction with external fields or with the presence of fluxes (like shear) in the surrounding fluid.

Second, we have shown how by considering the two body dynamics of Brownian Hard Spheres it is possible to develop an algorithm (the Approximate Green's function Dynamics AGD) that overcomes such problem and works equally well for a wide range of packing fractions and viscosities. To develop the AGD algorithm, we have solved the problem of the Langevin dynamics $\partial_t v = -\gamma v + \xi$ of a point particle in presence of a reflective wall by extending the classical Image Method solution for the over-damped Brownian dynamics $\partial_t x = \eta$ of a point particle in presence of a reflective wall (here ξ, η are noises). The AGD algorithm is Event Driven and considers fictive collisions between Hard Spheres. While it should be possible to take into account the polydispersity of a system by considering also effective masses in the fictive collisions as in [29], including shear or external fields in the AGD algorithm looks more complicated as it would require the solution of the particle - reflective wall problem with external fields/shear.

Both SBD and AGD simulations explore the canonical ensemble for Hard Spheres and therefore reproduce the correct equilibrium thermodynamics. They belong to the class of Asynchronous Event-Driven Particle Algorithms[40] and can be easily implemented by adapting existing codes for ED dynamics [36] or Brownian Dynamics [41] of Hard Spheres.

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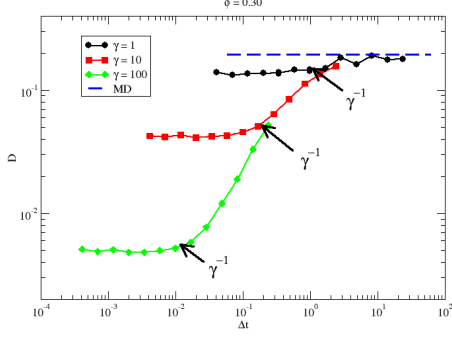


FIG. 1. Effect of the damping coefficient γ on the size of the simulation step Δt (all quantities in reduced units). The diffusion coefficient D from simulations is plotted versus the time-step size Δt for various γ 's. As expected, the system approaches the MD value for diffusion regardless of γ for $\Delta t \rightarrow \infty$. The “true” value of D is obtained for $\Delta t \rightarrow 0$. We observe at small Δt 's a plateau in the D vs Δt plot for $\Delta t \lesssim \gamma^{-1}$, signalling that the “true” value of D is approached. Results are presented for packing fraction $\phi = 0.30$; a completely analogous behaviour is found at a low packing fraction $\phi = 0.10$ and an high packing fraction $\phi = 0.45$.

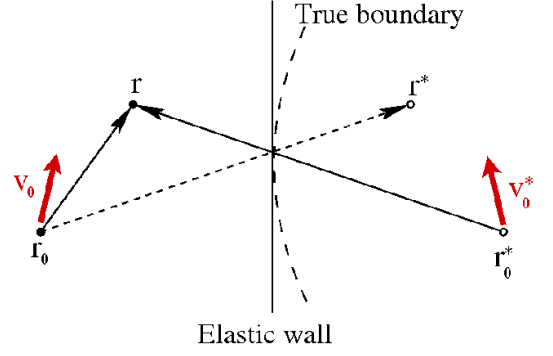


FIG. 2. A two body problem for hard spheres can be mapped into the problem of a point particle interacting with a larger sphere. When particles are very near, the problem further simplifies to the interaction of a Langevin particle with a reflective flat wall, whose solution can be derived by applying the Image Method to the Langevin equation. In fact, the Green function must zero inside the wall and must satisfy the no-flux boundary conditions at the wall. Combining the free Green function of the particle in its initial position and the free Green function of its image (with the normal-to-the-wall component of the initial velocity reflected) satisfies both Kramer' equations and reflective boundary conditions giving the correct solution.

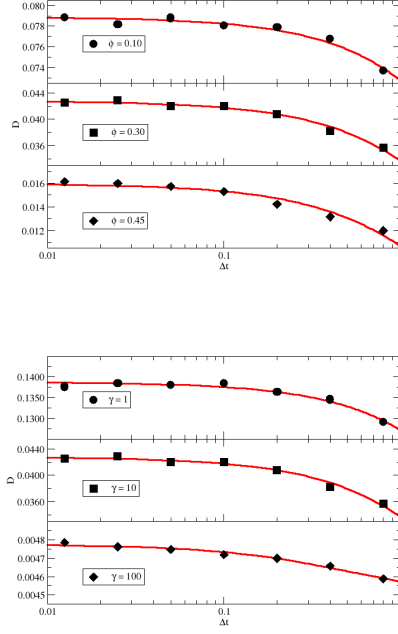


FIG. 3. Effects of packing fraction ϕ (upper panel) and of the damping coefficient γ (lower panel) on the time-step Δt for the AGF algorithm. All quantities in reduced units; thick lines are just a guide for the eye. Diffusion D is calculated averaging over 10 independent trajectories for 2000 particle systems; simulations are long at least 10 times the structural correlation time. In the upper panel, results are shown for $\phi = 0.10, 0.30, 0.45$ at fixed damping $\gamma = 10$. In the lower panel, results are shown for $\gamma = 1, 10, 100$ at fixed packing fraction $\phi = 0.30$. Notice that the estimated diffusion coefficient D has a small relative variation in the wide range of dampings γ s and packing fractions ϕ s analysed. As a rule of thumb, to estimate D with an accuracy much smaller than 1% time-step of order $\Delta t \sim 0.1$ are already enough.